9. Soil and Sediment Monitoring

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Introduction

Soil is weathered material, mainly composed of disintegrated rock and organic material, that is suitable for growing plants. Soil can contain pollutants originally released directly to the ground, to the air, or through liquid effluents. DOE guidance for environmental monitoring (U.S. Department of Energy 1991) states that soil should be sampled to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment and to estimate environmental radionuclide inventories. The guidance specifies that radionuclides in use at the facility as well as those that occur naturally should be monitored. Particulate radionuclides are of major interest in the LLNL soil monitoring program because airborne particulate releases are the most likely pathway for LLNL-induced soil contamination.

Sediments are defined, for the purposes of this chapter, as finely divided solid materials that have settled out of a liquid stream or standing water. To evaluate current conditions, LLNL samples recent sediments in storm drainage channels and the two arroyos on site. The accumulation of radioactive materials in sediment could lead to exposure of humans through ingestion of aquatic species, through sediment resuspension into drinking water supplies, or as an external radiation source (U.S. Department of Energy 1991). Note, however, that the Livermore site and Site 300 do not have habitats for aquatic species that are consumed by people, nor do they have surface drainage that directly feeds drinking water supplies.

Since 1971, surface soil sampling in the vicinity of the Livermore site and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity and to evaluate any increase in radioactivity that might have resulted from LLNL operations. These samples have been analyzed for plutonium and gamma-emitting radionuclides, such as depleted uranium, which is occasionally used in high-explosives tests at Site 300. The inclusion of other gamma-emitting naturally occurring nuclides (^{40}K, ^{232}Th, and ^{235}U) and the long-lived fission product ^{137}Cs provides background information and baseline data on global fallout.

Similarly, sediment samples have been collected from selected arroyos and other drainage areas at and around the Livermore site since 1988; these locations largely coincide with selected storm water sampling locations. The number of sediment sampling locations was reduced in 1994 to correspond to reductions in storm water sampling locations. In addition, in 1991, LLNL began analyzing

surface soil samples for beryllium, a potentially toxic metal used at both the Livermore site and Site 300.

Location maps for soil and sediment sampling conducted during 1994 are provided in **Figures 9-1** through **9-3**. The locations were selected to represent background concentrations (distant locations unlikely to be affected by LLNL operations) as well as areas where there is the potential to be affected by LLNL operations. Areas with known contaminants, such as the Livermore Water Reclamation Plant (LWRP), are also sampled. In general, Site 300 soil sampling locations were established around firing tables and other areas of potential soil contamination. Arroyo and drainage channel sediment sampling locations were chosen to coincide with major Livermore-site storm water drainages. All soil and sediment sampling locations have permanent location markers for reference.

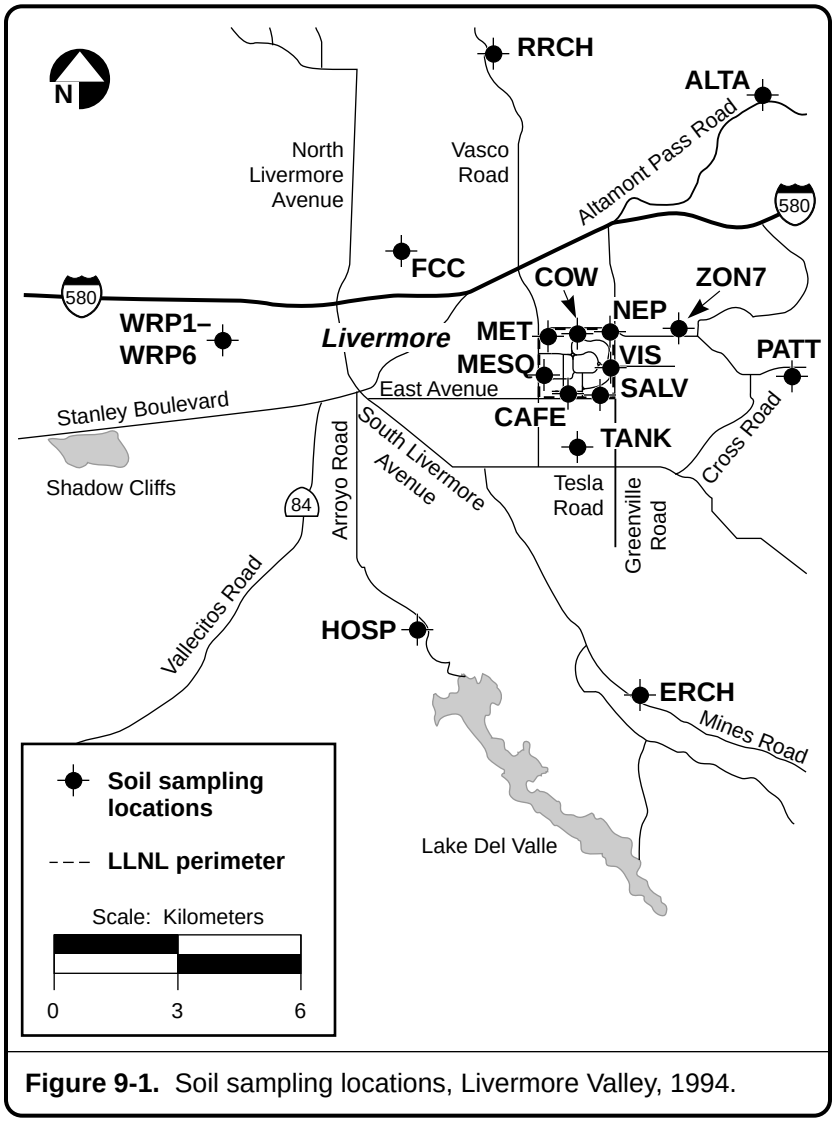

Figure 9-1. Soil sampling locations, Livermore Valley, 1994.

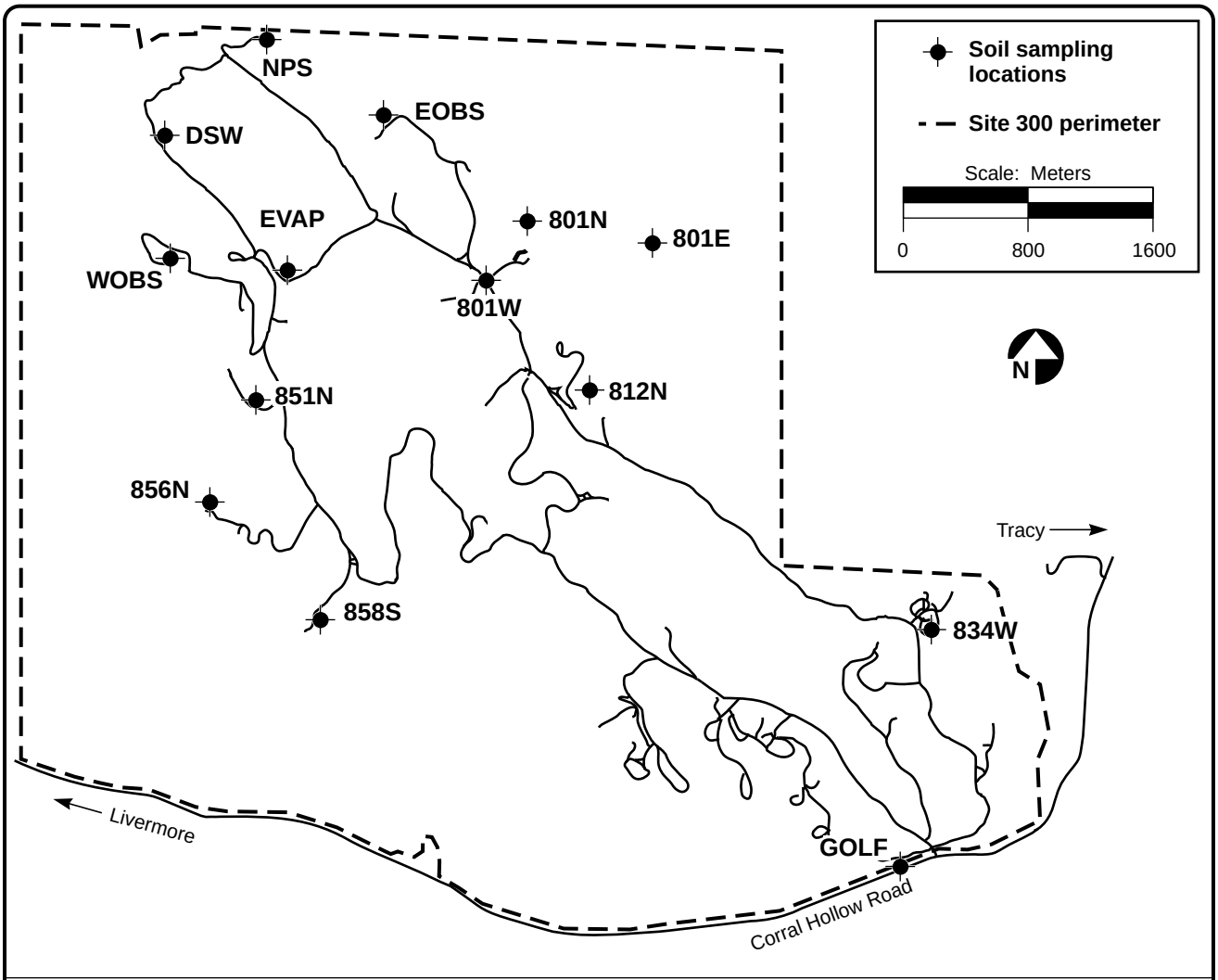

Figure 9-2. Site 300 soil sampling locations, 1994.

Methods

Soil and sediment sampling is conducted according to written, standardized procedures (Tate et al. 1995, Appendix A). Soil samples are collected from undisturbed areas near the permanent sampling location marker. These areas generally are level, free of rocks, and are unsheltered by trees or buildings. All samples are collected from the top 5 centimeters of soil because surface deposition from the air is the primary pathway for potential contamination. Quality control samples are submitted with each batch of soil samples. At locations chosen for duplicate sampling, two identical samples are collected.

Samples of recent sediment are collected annually from drainages at and around the Livermore site after the cessation of spring runoff. For 1994, samples were

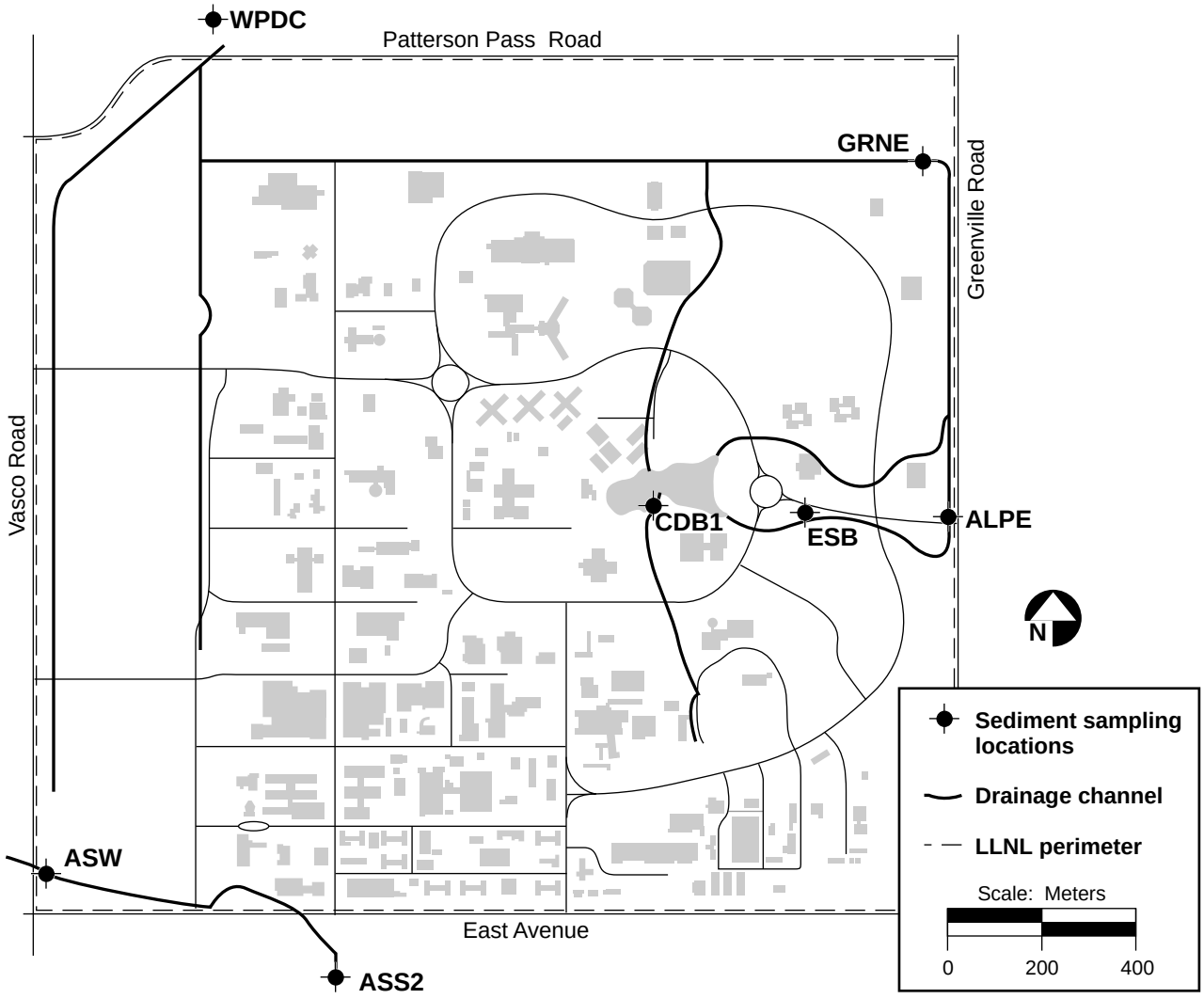

Figure 9-3. Arroyo and drainage basin sediment sampling locations, 1994.

analyzed for radionuclides and beryllium. Critical evaluation of the sediment monitoring program for heavy metals and organic compounds in 1994 did not yield sufficient evidence of contamination to warrant further yearly sampling (Tate et al. 1995). LLNL staff will continue to explore the need for sediment sampling for heavy metals and organic compounds as new regulations are developed or as LLNL operations change.

Soils and sediment samples are delivered on the day of collection to LLNL's Radiation Analytical Sciences (RAS) laboratory for analyses. Soil samples are dried, ground, sieved, and blended. The plutonium content of a sample aliquot is determined by alpha spectroscopy (Hall and Edwards 1994). Other sample aliquots (300 grams) are analyzed for more than 150 radionuclides by gamma

spectroscopy, using a high-purity germanium (HPGe) detector (Hall and Edwards 1994). The 10-gram subsamples for beryllium analyses are sent to a contract analytical laboratory and are analyzed by graphite-furnace atomic absorption spectroscopy. For samples collected for tritium analyses, RAS uses freeze-drying techniques to recover water from the samples, and determines the tritium content of the water by liquid-scintillation counting. Chain-of-custody procedures are followed throughout the sampling, delivery, and analytical processes.

Livermore Valley Results

Table 9-1 presents summary data on the concentrations of $^{239+240}$Pu, ^{40}K, ^{60}Co, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U, in surface soils from the Livermore Valley sampling locations. The complete data for 1994 soils and sediment sampling is presented in Table 9-1, Volume 2, of this report. The concentrations and distributions of all observed radionuclides in soil for 1994 are within the ranges reported in previous years and generally reflect worldwide fallout and naturally occurring concentrations. The ratio of ^{235}U to ^{238}U reflects the natural ratio of 0.7%; however, there is uncertainty in the ^{235}U / ^{238}U ratio due to the difficulty in measuring ^{238}U by gamma spectroscopy.

As in 1991, $^{239+240}$Pu was detected at background levels—0.22×10^{-3} Bq / g (6.1×10^{-3} pCi / g)—at location ZON7. Since 1973, soil samples in this area have generally shown $^{239+240}$Pu values that are higher than background. The slightly higher values at the Livermore site have been attributed to historic operations, which included the operation of solar evaporators for plutonium-containing liquid waste in the southeast quadrant (Silver et al. 1974). LLNL no longer operates the solar evaporators or any other open air treatment of plutonium-containing waste. Nonetheless, $^{239+240}$Pu from historic operations is carried off site by resuspension of soil and other particles by wind. Similarly, elevated levels of $^{239+240}$Pu, resulting from an estimated 1.2×10^9 Bq (32 mCi) plutonium release to the sewer in 1967 and first observed in soils near LWRP during the early 1970s, again were detected at LWRP sampling locations.

Of all the factors that could effect the measured activity of $^{239+240}$Pu in soils, the particle size of the $^{239+240}$Pu levels may be the most significant. The radioactivity of a particle is proportional to the third power of the diameter of the particle; e.g., a particle 10 times bigger than a second particle is 1,000 times as radioactive. Using the equation from Sill (1971)—Activity (Bq) = 0.01202 * Number of particles * (Diameter of particles)^3—and, for the sake of discussion, assuming one particle per gram of soil, the geometric mean of historical plutonium results from the Livermore Valley (1.17 × 10–4 Bq / g) suggests that the average particle size is

Table 9-1. Summary of soils and sediment analytical data, 1994.

Analyte and Location	Detection Frequency	Median	IQR[a]	Maximum
$^{239+240}$Pu (10^{-3} Bq/dry g)				
Livermore Valley soils	15/15	0.09	0.20	0.64
LWRP soils	6/6	4.4	9.6	38
Livermore site sediments[b]	6/6	0.10	1.3	2.1
Site 300 soils	14/14	0.11	0.08	0.25
^{137}Cs (10^{-3} Bq/dry g)				
Livermore Valley soils	14/15	3.1	1.6	10
LWRP soils	6/6	3.6	3.8	5.9
Livermore site sediments[b]	5/6	0.93	0.7	1.5
Site 300 soils	14/14	4.5	3.1	9.3
^{40}K (Bq/dry g)				
Livermore Valley soils	15/15	0.459	0.11	0.599
LWRP soils	6/6	0.427	0.037	0.455
Livermore site sediments[b]	6/6	0.481	0.053	0.492
Site 300 soils	14/14	0.498	0.12	0.662
^{232}Th (μg/dry g)[c]				
Livermore Valley soils	15/15	6.5	1.4	8.4
LWRP soils	6/6	6.8	0.6	7.7
Livermore site sediments[b]	6/6	5.9	1.3	8.4
Site 300 soils	14/14	9.6	1.3	44
^{235}U (μg/dry g)[d]				
Livermore Valley soils	12/15	0.025	0.009	0.032
LWRP soils	6/6	0.027	0.012	0.036
Livermore site sediments[b]	4/6	<0.019	—[e]	0.028
Site 300 soils	12/14	0.029	0.014	1.8
^{238}U (μg/dry g)[f]				
Livermore Valley soils	6/15	2.2	—[e]	3.8
LWRP soils	5/6	2.7	1.0	3.5
Livermore site sediments[b]	2/6	<2.0	—[e]	3.4
Site 300 soils	9/14	3.4	6.4	870

Table 9-1. Summary of soils and sediment analytical data, 1994 (concluded).

Analyte and Location	Detection Frequency	Median	IQR[a]	Maximum
^{3}H (Bq/L extracted water)[g]				
Livermore site sediments	5/6	3.1	9.4	20
^{60}Co (10^{-3} Bq/dry g)[h]				
LWRP soils	2/6	<0.1	—[e]	0.5
Be (mg/kg)[i]				
Livermore Valley soils	15/15	0.50	0.28	1.2
LWRP soils	6/6	0.64	0.25	0.80
Site 300 soils	14/14	1.5	0.65	42

a IQR = interquartile range.

b Location WPDC could not be sampled because water was flowing through the drainage channel.

c Thorium-232 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 247.3, and pCi/dry g can be determined by dividing by 9.15.

d Uranium-235 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 12.5, and pCi/dry g can be determined by dividing by 0.463.

e IQR could not be calculated.

f Uranium-238 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 80.3, and pCi/dry g can be determined by dividing by 2.97.

g Tritium (^{3}H) analysis is only conducted on sediment samples.

h Cobalt-60 is only detected in LWRP soil samples.

i Beryllium analysis is only conducted on soils samples; the analysis is a chemical, not a radiochemical analysis.

0.21 micron. A particle twice as large as the average particle (0.41 micron) would result in a measured activity of 9.4×10^{-4} Bq / g, eight times the average; in contrast, it would take eight 0.21-micron particles to result in the same measured activity. Consequently, the presence of a larger than average particle will cause a disproportionately large analytical result.

Historical plots of average $^{239+240}$Pu concentrations in soil in the Livermore Valley, at Site 300, and at LWRP are shown in **Figure 9-4**. Livermore Valley and Site 300 concentrations have remained relatively constant over the past ten years and generally are indicative of worldwide fallout (locations VIS and ZON7 at the Livermore site show activities greater than background). Greater variability in $^{239+240}$Pu is seen at LWRP. Six samples are being evaluated to determine the median at LWRP. The $^{239+240}$Pu is likely to be present in discrete particles, so the presence or absence of the particles will dominate the measured $^{239+240}$Pu in any given sample.

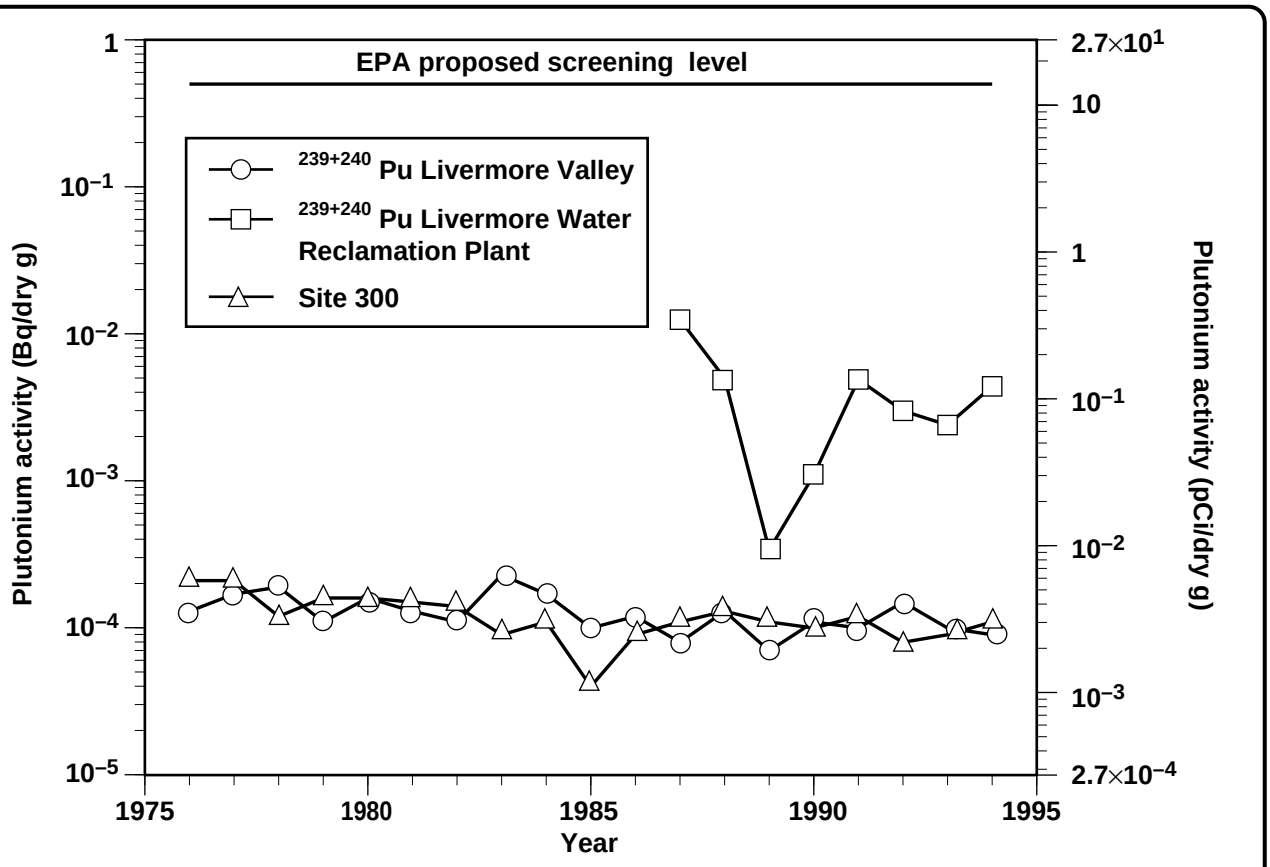

Figure 9-4. Median plutonium-239+240 activities on surface soils, 1976 to 1994.

As in 1991 to 1993, low levels of ^{60}Co were detected at the LWRP. While there is ^{60}Co in use at the Livermore site, it is only present in gram quantities in three facilities (Buildings 151, 194, and 514) or in sealed sources. Low levels of ^{60}Co, on the order of 0.0037 Bq / g (0.1 pCi / g), have also been detected intermittently in sewage sludge samples. If the Livermore site were the source of ^{60}Co, this activity of ^{60}Co in the sludge would translate into about 1.5×10^{-6} Bq / mL (40×10^{-6} pCi / mL) in the effluent leaving the site, which is below the detection limits of current analytical methods. This level is also well below the DOE effluent limit of 0.925 Bq / mL (25 pCi / mL). The reader should note that LLNL is not the only contributor to the waste stream that arrives at the LWRP and that ^{60}Co is used in a variety of medical, technical, and research applications. It is not possible to determine if LLNL is the source of ^{60}Co at LWRP. However, it can be concluded that LLNL controls on the release of ^{60}Co are sufficient to ensure that LLNL activities do not adversely affect LWRP operations.

Table 9-1 shows data on the concentrations of beryllium in surface soils from Livermore Valley sampling locations. Beryllium levels in soil samples from the Livermore Valley were comparable to the normal range of background

concentrations (Wilber 1980). Beryllium analysis for Livermore Valley soils will be discontinued in 1995. The few LLNL operations that use beryllium are HEPA filtered. In addition, sampling data to date have shown no evidence of beryllium contamination in the Livermore Valley (Tate et al. 1995). Should beryllium usage change, LLNL's environmental monitoring staff would reevaluate the need for beryllium monitoring in soils.

Table 9-1 presents summary data on radionuclides detected in the sediment samples; a complete presentation of 1994 sediment data is found in Table 9-1, Volume 2, of this report. The levels of $^{239+240}$Pu were generally at background concentrations, reflective of worldwide fallout. The higher values at CDB1 and ESB may be attributed to historic activities in the southeast quadrant at LLNL; these locations are both in drainages for that area. Most other radionuclides were detected at levels similar to those reported from 1988 through 1991: ^{137}Cs, a fission product, was found at worldwide background concentrations; and ^{40}K, ^{232}Th, ^{235}U, and ^{238}U—naturally occurring radionuclides—were detected at background concentrations. Tritium concentrations were below those reported from 1988 through 1992, but above those for 1993. Median tritium values are shown in **Figure 9-5** and show a general decline since measurement began. In 1993, the sediment sampling procedure was changed so that samples were collected 5 cm deep, rather than 15 cm deep; both 1993 and 1994 samples were

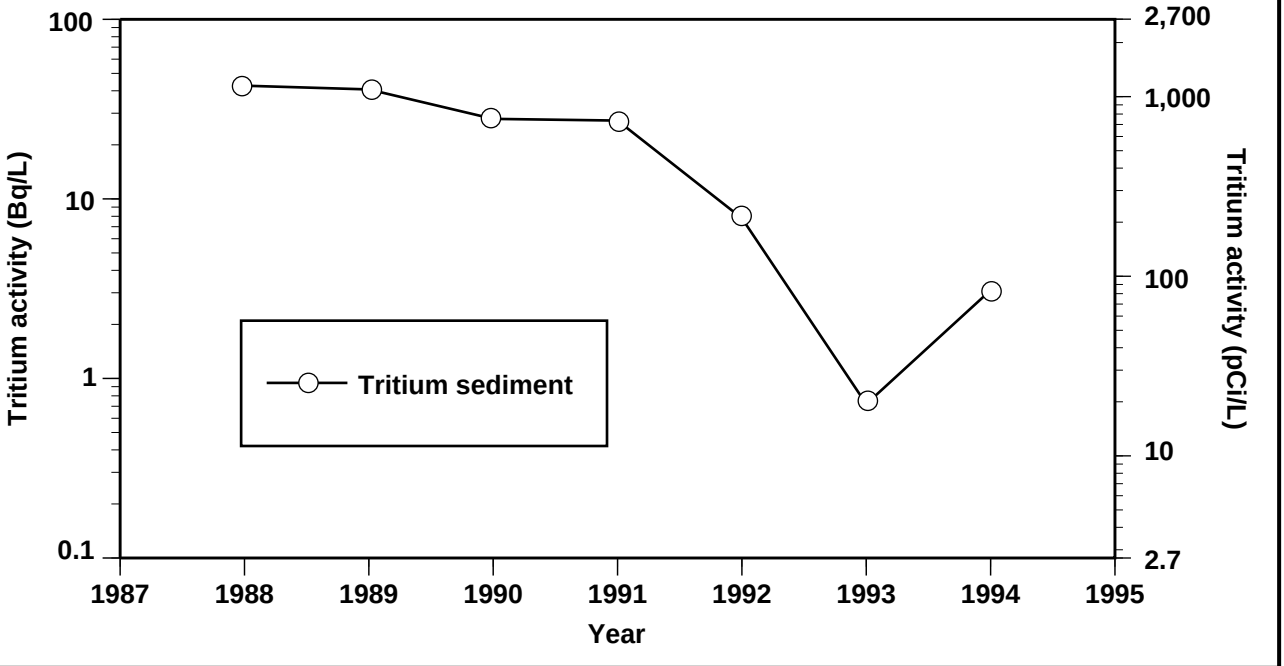

Figure 9-5. Median tritium concentrations in sediments (Bq/L of recovered water), 1988 to 1994.

collected at the shallower depth. The effect of the change in sampling depth, if any, on measured activities is not clear; nonetheless, it appears from the **Figure 9-5** that tritium values for 1993 were unusually low compared to all other years. Tritium in sediments will continue to be evaluated.

Site 300 Results

Table 9-1 presents summary data on the concentrations of $^{239+240}Pu$, ^{40}K, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U in soil from the Site 300 sampling locations; a complete presentation of 1994 soils data for Site 300 is found in Table 9-1, Volume 2, of this report. The concentrations and distributions of all observed radionuclides in Site 300 soil for 1994 lie within the ranges reported in all years since monitoring began, and, with one exception discussed below, reflect naturally occurring concentrations. The ratio of ^{235}U to ^{238}U reflects the natural ratio of 0.7%.

Historical trends of ^{238}U concentrations from both the Livermore Valley and Site 300 are shown in **Figure 9-6**. Median values have remained relatively constant for both places. The highest values at Site 300 are caused by the use of depleted uranium in high-explosive tests.

One sample from a region near a firing table (812N) had substantially higher than background concentrations of ^{238}U and beryllium. To investigate the elevated ^{238}U and beryllium result at 812N, LLNL personnel resampled the original sampling location as well as four additional locations about 5 meters north, south, east, and west of the original 1994 sampling location. The results of this investigation are shown in **Table 9-2.** The highest value of 870 μg/g of ^{238}U is the same order of magnitude as a high ^{238}U value found at that location in 1988 (570 μg/g) (**Figure 9-6**). The $^{235}U/^{238}U$ ratios, at 0.2%, confirm the presence of depleted uranium; the ratio in naturally occurring material is 0.7%. Beryllium analyses have only been conducted since 1991, so there is less historic data for comparison of beryllium results. The 1994 results for beryllium at 812N are well above the previous highest beryllium result in 1992, which was 2.5 mg/kg at location 801N. The samples for the 812 area also showed elevated levels of ^{232}Th; these 1994 results are similar to, but higher than, ^{232}Th results for environmental samples in the 812 area for 1987 (20 μg/g) and 1988 (18 μg/g). As with beryllium, there is limited historic data for ^{232}Th at Site 300; the ^{232}Th results have only been reported since 1987. The results for beryllium and ^{238}U in the Building 812 area are confirmed in previous reports published as part of the Site 300 Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) restoration project (Lamarre et al. 1989c; Webster-Scholten 1994). Further investigation of this area is planned during CERCLA restoration.

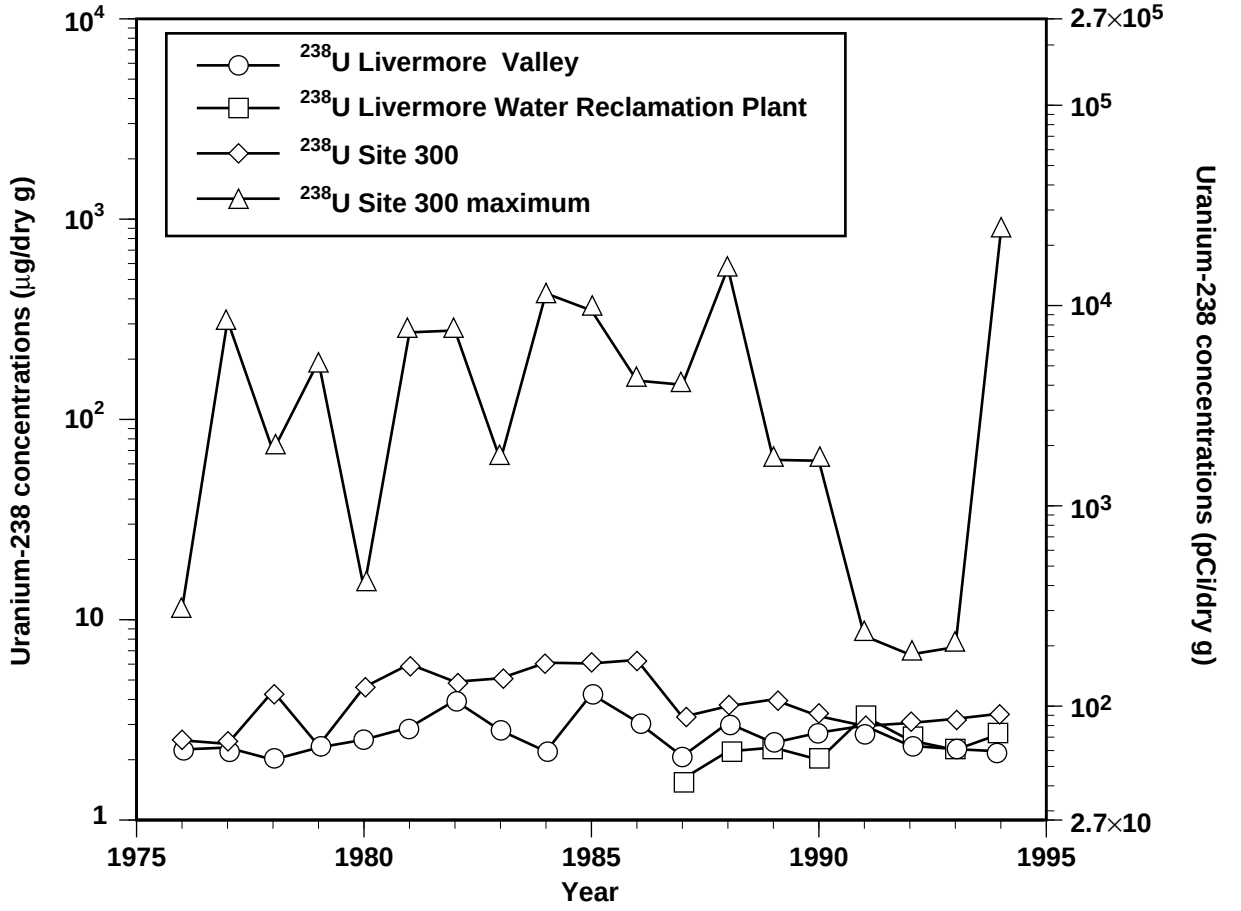

Figure 9-6. Median uranium-238 concentrations in surface soils, 1976 to 1994.

Table 9-2. Sample results for location 812N for 1994.

	Beryllium (mg/kg)	^{238}U (μg/dry g)	^{235}U (μg/dry g)	Ratio ^{235}U/^{238}U	^{232}Th (μg/dry g)
Original sample	42	870	1.8	0.002	44
Rerun of original	240	780	1.8	0.002	—[a]
Resample					
Original location	23	260	0.56	0.002	26
East of original	9.3	400	0.89	0.002	35
North of original	11	420	0.89	0.002	41
South of original	38	470	1.0	0.002	33
West of original	14	420	0.9	0.002	32

[a] Sample not analyzed for ^{232}Th.

Environmental Impact

This section discusses the environmental impacts at the Livermore site and Site 300.

Livermore Site

Routine soil and sediment sample analyses indicate that the impact of LLNL operations on these media in 1994 has not changed from previous years and remains insignificant. Most analytes of interest or concern were detected at background concentrations, in trace amounts, or could not be measured above detection limits.

The highest value of 38×10^{-3} Bq/g (1.0 pCi/g) for $^{239+240}$Pu measured at LWRP during 1994 represents 7.6% of the proposed EPA surface soil screening level of 7,400 Bq/m^2 (0.2 μCi/m^2), or 0.5 Bq/g (13 pCi/g), assuming average Livermore Valley soil densities of 1.5 g/cm^3 and a potential resuspension depth of 1.0 cm. (Areas that do not exceed the screening level are generally said to be in compliance and need no further investigation for possible remediation.) The proposed EPA screening level for surface soil contamination was derived from conservative assumptions and mathematical models that considered both the inhalation and ingestion pathways (42 Federal Register 230 1977; U.S. Environmental Protection Agency 1977, 1978). Statistical analysis shows that all LWRP $^{239+240}$Pu soils data are lognormally distributed, and there is no general increase or decrease in $^{239+240}$Pu values with time. Moreover, all measured concentrations, regardless of location and year, have been a small fraction of the proposed EPA screening level, which is shown in **Figure 9-4** for comparison.

In addition, evaluation of the soils and sediment monitoring program (Tate et al. 1995) has shown that LLNL nonradiological impacts on sediment and airborne releases of beryllium are not sufficient to warrant continued sampling. Sampling of soils for radiological materials will continue on an annual basis.

Site 300

With the exception of elevated concentrations of ^{238}U, Be, and possibly ^{232}Th, at location 812N, the concentrations of radionuclides and beryllium observed in soil samples collected at Site 300 are representative of background or naturally occurring levels. In 1988, contaminated gravel from the firing table at Building 812 was removed to on-site landfills, and measured values for samples from this location have generally not exhibited elevated levels of ^{238}U and beryllium. The elevated results for ^{238}U and beryllium indicate that areas outside the firing table may be contaminated by firing table debris. The investigation planned as part of the Site 300 CERCLA restoration efforts will clarify the nature and extent of the contamination in this area.

Special Studies

LLNL was involved with two special studies of soils in 1994: Plutonium in the Soil in the Southeast Quadrant of Livermore Site and Plutonium in the Soil in Big Trees Park, Livermore. The results of these studies follow.

Plutonium in Soil, Southeast Quadrant of Livermore Site

From 1962 to 1976, solar evaporation trays were located in the southeast quadrant of LLNL. The trays were approximately 6 meters × 6 meters × 3 meters deep, constructed of concrete coated with polyamide epoxy paint, and lined with polyvinylchloride or polyethylene liners. Plutonium-containing liquid waste was put in these trays to reduce by evaporation the total volume of disposable waste (Buerer 1983).

In 1991, in response to a Tiger Team comment, 195 surface soil samples were collected and analyzed for plutonium in the southeast quadrant of LLNL. None of the samples were above the interim EPA guidance for Superfund remediation for commercial/industrial sites (0.51 Bq/g or 13.7 pCi/g). The highest level detected was 0.11 Bq/g (3 pCi/g). In 1993, EPA decided to resample the areas with levels above the global fallout for further conformation and to sample locations to the west of the 1991 sampling locations to assure the boundary of the area of interest had been appropriately set.

The highest plutonium values in the 1993 study were 0.32 Bq/g (8.6 pCi/g) at a depth of 0.01 meters and 0.45 Bq/g (12.2 pCi/g) at a depth of 0.05 meters. These values are higher than the highest value found in the 1991 study. Comparison of the 1993 data with the data from 1991 and a previous study from 1974, shows that plutonium activities have remained substantially the same (Gallegos et al. 1994).

Plutonium in Soil, Big Trees Park, Livermore

During the 1993 EPA investigation of plutonium in soils in the southeast quadrant of the Livermore site, EPA personnel collected a soil sample at Big Trees Park in Livermore to obtain a background sample. This soil sample showed plutonium at a concentration higher than what is expected from global fallout for this region. The park was resampled by EPA, LLNL, and the California Department of Health Services (DHS) in 1995. The results confirmed the finding of plutonium, with all the results below the EPA's health protective screening level for residential exposure. The EPA and DHS concur that there is no regulatory concern or significant impact on human health and the environment.